

Estimating the compressibility of osmium from recent measurements of Ir-Os alloys under high pressure

N. V. Sarlis and E. S. Skordas,
 Department of Solid State Physics and
 Solid Earth Physics Institute
 Faculty of Physics, School of Science,
 National and Kapodistrian University of Athens,
 Panepistimiopolis, Zografos 157 84,
 Athens, Greece

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Abstract

Several fcc- and hcp-structured Ir-Os alloys have been recently studied up to 30 GPa at room temperature by means of synchrotron-based X-ray powder diffraction in diamond anvil cells. Using their bulk moduli -which increase with increasing osmium content showing a deviation from linearity- and after employing a thermodynamical model it was concluded that the bulk modulus for osmium is slightly smaller than that for diamond. Here, a similar conclusion is obtained upon employing an alternative model, thus strengthening the conclusion that osmium is the densest but not the most incompressible element. This is particularly interesting for Earth Sciences since it may be of key importance towards clarifying the anomalous elastic properties of the Earth's core.

1 Introduction

Heavy transition metals such as Ta, W, Re, Os and Ir have exceptional mechanical, thermal and chemical stabilities. Their very low compressibilities κ make them essential subjects for high pressure studies. The compressibility of materials, in general, is of major importance because of its correlation to material strength, chemical bonding, and electronic structure. Strongly bonded materials have short interatomic distances and correspondingly strong repulsive forces, resulting in high bulk moduli $B(= 1/\kappa)$. In this regard, the finding by *Cynn et al.* (2002) that the B value for Os at ambient conditions is $B = 462(12)$ GPa, i.e., larger than that for diamond ($B = 443$ GPa, see *Occelli et al.* (2003)), attracted major interest. Such a large B value has implications for the nature

of the metallic bond in Os, where the bonding electrons are delocalized compared to the covalently bonded diamond in which they are localized. Later studies, however, came out with markedly lower B values for Os. In particular, values around 400GPa were reported by *Occelli et al.* (2003), *Kenichi* (2004) and *Godwal et al.* (2012), while *Voronin et al.* (2005) found $B = 435(19)$ GPa and *Armentrout and Kavner* (2010) $B = 421(3)$ GPa. Hence, controversial values concerning the bulk modulus for Os have been published to date. This is challenging in view of the following: Osmium is the densest element at ambient conditions, thus if its corresponding B value exceeds that of diamond (as reported by *Cynn et al.* (2002)), this would reflect that Os is also the most incompressible element. In addition, osmium is of particular importance in Earth Sciences because it provides key information about the properties of iron and its alloys making up Earth's core: The stability of hcp osmium is similar to that of hcp iron, and a systematic characterization of these analog materials will help towards understanding the anomalous properties of Earth's core (*Morelli et al.*, 1986; *Creager*, 1992). The melting point of osmium has been reported to be 3306K (*Reiswig and Dickinson*, 1964; *Okamoto*, 1994; *Vacher et al.*, 1954; *Geballe et al.*, 1963).

An interesting study of the compressibility of Ir-Os alloys under high pressure just appeared by *Yusenko et al.* (2015). As they noticed phase stability in the Ir-Os system has importance for the genesis of the natural Ir-Os-Ru and other pure platinum group based metallic minerals which show complex nature and non-equilibrium character at ambient conditions due to their formation under high-pressure from poly-component melt in Earth's mantle (*Bird and Bassett*, 1980; *Fonseca et al.*, 2012; *Weinberger et al.*, 2008). They prepared several fcc- and hcp- structured Ir-Os alloys and the measurement of the corresponding atomic volumes at ambient conditions using powder X-ray diffraction showed an almost linear dependence as a function of composition. These alloys were studied up to $P = 30$ GPa at room temperature by means of synchrotron-based X-ray powder diffraction in diamond anvil cells and their bulk moduli were found to increase with increasing osmium content showing a deviation from linearity. This concentration dependence of bulk moduli was shown by *Yusenko et al.* (2015) to be satisfactorily described by a thermodynamic model developed by *Varotsos* (1980a). According to this model the bulk moduli $B_0(x_{\text{Os}})$ as a function of the atomic fraction of osmium, x_{Os} , is given by

$$B_0(x_{\text{Os}}) = B_{\text{Ir}} \left[\frac{1 + x_{\text{Os}} \left(\frac{V_{\text{Os}}}{V_{\text{Ir}}} - 1 \right)}{1 + x_{\text{Os}} \left(\frac{B_{\text{Ir}} V_{\text{Os}}}{B_{\text{Os}} V_{\text{Ir}}} - 1 \right)} \right], \quad (1)$$

where B_{Ir} and B_{Os} are bulk moduli and V_{Ir} and V_{Os} are atomic volumes at ambient pressure of pure Ir and Os, respectively. *Yusenko et al.* (2015), upon using their measured bulk moduli of the Ir-Os alloys and employing Eq.(1), found the values 354(2)GPa and 442(4)GPa for pure Ir and Os, respectively. In particular, *Yusenko et al.* (2015) concluded that the bulk modulus for pure Os is slightly smaller than that of the diamond. It is the scope of the present short

paper to investigate the validity of this conclusion of *Yusenko et al.* (2015) by using an alternative model which interrelates the bulk modulus of an alloy with the bulk moduli of its pure constituents. The basic idea of this model is that when replacing an atom of a host crystal by a “foreign” atom, the corresponding volume variation can be considered as a “defect volume”.

2 The alternative model for the compressibility of a solid solution

Let us explain how the compressibility κ of a solid solution A_xB_{1-x} is interrelated with compressibilities of the two end members A and B by following *Varotsos and Alexopoulos* (1986) and *Skordas* (2012). We call the two end members A and B as pure components 1 and 2, respectively, and label v_1 the volume per atom of the pure component 1 and v_2 the volume per atom of the pure component 2. Let V_1 and V_2 denote the corresponding molar volumes, i.e. $V_1 = Nv_1$ and $V_2 = Nv_2$ (where N stands for Avogadro’s number) and assume that $v_1 < v_2$. We now define a “defect volume” (*Varotsos and Alexopoulos*, 1986; *Skordas*, 2012) as the increase of the volume V_1 , if one atom of type 1 is replaced by one atom of type 2. It is evident that the addition of one “atom” of type 2 to a crystal containing atoms of type 1 will increase its volume by $v_2 - v_1$ (see pp.325 and 326 of *Varotsos and Alexopoulos* (1986) as well as *Skordas* (2012)). Assuming that v^d is independent of composition, the volume V_{N+n} of a crystal containing N atoms of type 1 and n atoms of type 2 can be written as

$$V_{N+n} = Nv_1 + n(v^d + v_1) \iff V_{N+n} = \left[1 + \frac{n}{N}\right] V_1 + nv^d. \quad (2)$$

The molar fraction x is connected to n/N by (see Eq.(12.5) on page 328 of *Varotsos and Alexopoulos* (1986))

$$\frac{n}{N} = \frac{x}{1-x}. \quad (3)$$

The compressibility κ of the solid solution (as well as its bulk modulus $B = 1/\kappa$) can be found by differentiating Eq.(2) with respect to pressure, which finally gives:

$$\kappa V_{N+n} = \kappa_1 V_1 + \frac{n}{N} [\kappa^d N v^d + \kappa_1 V_1], \quad (4)$$

where κ^d denotes the compressibility of the volume v^d , defined as

$$\kappa^d \equiv \frac{1}{B^d} = -\frac{1}{v^d} \left(\frac{\partial v^d}{\partial P} \right)_T. \quad (5)$$

The “defect volume” v^d can be approximated by (see p.342 of *Varotsos and Alexopoulos* (1986))

$$v^d = \frac{V_2 - V_1}{N} = v_2 - v_1. \quad (6)$$

Obviously V_{N+n} can be obtained versus the composition by means of Eq.(2) when considering also Eq.(6). Then, the compressibility κ can be studied versus the composition from Eq.(4) by assuming -to a first approximation- that the compressibility κ^d is independent of composition. A rough estimation of κ^d can be made by employing a thermodynamical model (Varotsos and Alexopoulos, 1977; Varotsos et al., 1978) for the formation and migration of the defects in solids (cf. the replacement of a host atom with a “foreign” one can be considered in general as a defect (Varotsos, 1974; Varotsos and Mourikis, 1974; Varotsos and Miliotis, 1974)). This model has been successfully applied to various categories of solids including diamond (Varotsos, 2007), oxides (Chroneos and Vovk, 2015a), semiconductors (Chroneos and Vovk, 2015b), silicates (Zhang and Shan, 2015) metals (Alexopoulos and Varotsos, 1981; Varotsos and Alexopoulos, 1980a), ionic crystals (Varotsos and Alexopoulos, 1980b), fluorides (Varotsos and Alexopoulos, 1981, 1980c; Varotsos et al., 1985; Varotsos, 2008), mixed alkali halides (Varotsos, 1980b, 1981; Varotsos and Alexopoulos, 1980d; Varotsos, 1978) as well as complex ionic materials under uniaxial stress that emit electric signals before fracture (Varotsos et al., 1999, 1992), thus explaining the signals detected before major earthquakes (Varotsos, 2006; Varotsos et al., 2009, 2008; Skordas et al., 2010; Sarlis et al., 2010). Within the frame of this thermodynamical model, which states that the defect Gibbs energy g is proportional to the bulk modulus as well as to the mean volume per atom, we first obtain the defect volume $v=[(\partial g/\partial P)_T]$, and therefrom the compressibility κ^d (see Eq.(8.31) in p.156 of Varotsos and Alexopoulos (1986)):

$$\kappa^d = \frac{1}{B} - \frac{\left(\frac{\partial^2 B}{\partial P^2}\right)_T}{\left(\frac{\partial B}{\partial P}\right)_T - 1}. \quad (7)$$

Since $\left(\frac{\partial^2 B}{\partial P^2}\right)_T$ is negative (see p.157 of Varotsos and Alexopoulos (1986)), Eq.(7) indicates that $\kappa^d > \frac{1}{B}$, i.e., $B > B^d$.

3 Application of the alternative model to Ir-Os alloys

Equation (4) can be alternatively written as

$$\left(\frac{V_{N+n}}{N+n}\right) \frac{1}{B} = \frac{V_1}{N} \frac{1}{B_1} + \frac{v^d}{B^d} \left(\frac{n}{N+n}\right) \quad (8)$$

where $\frac{V_{N+n}}{N+n}$ for the alloys denotes the mean volume per atom given in Table 3 of Yusenko et al. (2015). This equation reveals that when plotting $\left(\frac{V_{N+n}}{N+n}\right) \frac{1}{B}$ versus $\frac{n}{N+n}$ ($= x$, see Eq.(3)) and making a least square fitting to a straight line the slope leads to $\frac{v^d}{B^d}$ -from which B^d is calculated since v^d may be approximated

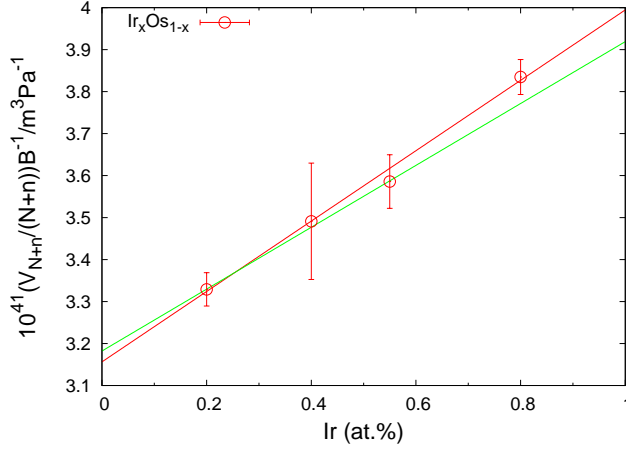


Figure 1: The composition dependence of the quantity $\left(\frac{V_{N+n}}{N+n}\right)\frac{1}{B}$ for the Ir_xOs_{1-x} alloys studied by *Yusenko et al.* (2015). The red straight line corresponds to all four alloys (red circles) according to Eq.(8), while in the green straight line the highest Ir concentration $x = 0.80$ is not considered (see the text).

by means of Eq.(6)- and the intercept results in $\frac{V_1}{N}\frac{1}{B_1}$ from which the bulk modulus B_1 of the pure component 1 is determined.

We follow the description of the previous Section and, for the sake of convenience, we assume as pure components 1 and 2 the elements Os and Ir (since the volume per atom in Os is smaller than that in Ir), thus we consider the alloys Ir_xOs_{1-x} . *Yusenko et al.* (2015) studied four compositions, i.e., $x = 0.20, 0.40, 0.55$ and 0.80 , and in their Table 3 reported the following bulk moduli data $B = 420(5), 403(16), 393(7)$, and $368(4)$ GPa, respectively. The mean volumes per atom for the two pure components are (*Yusenko et al.*, 2015) $v_1 = 13.9825(1) \times 10^{-30}m^3$ and $v_2 = 14.1556(1) \times 10^{-30}m^3$ and hence $v^d = 0.1731(1) \times 10^{-30}m^3$. Using these data, we now plot in Fig.1 with open red circles the values of $\left(\frac{V_{N+n}}{N+n}\right)\frac{1}{B}$ versus $\frac{n}{N+n}(=x)$ and make a weighted least squares fit (see pp. 659-660 of *Press et al.* (1992)).

The following results have been obtained: When employing all four alloys studied by *Yusenko et al.* (2015) we deduce the red straight line, the slope of which leads to $B^d = 20.6(2.3)$ GPa whereas its intercept to the bulk modulus of Os $B_1 = 443(8)$ GPa and the corresponding ordinate for $x = 1$ to the bulk modulus of Ir, $B_2 = 354(10)$ GPa. If we do not consider the alloy with the highest Ir concentration, i.e., the one with $x = 0.80$ (for which the concept of the “defects” introduced by Ir when considering Os matrix may not be fully justified) and restrict ourselves to the three alloys having the lower x values, i.e., $x = 0.20, 0.40$, and 0.55 , we deduce the green straight line from which slightly different values are obtained. In particular, the slope leads to $B^d =$

23(7)GPa, its intercept to $B_1 = 439(10)$ GPa and the value $x = 1$ corresponds to $B_2 = 361(21)$ GPa. Thus, our results of the application of the alternative model to $\text{Ir}_x\text{Os}_{1-x}$ alloys could be summarized as follows: The bulk modulus B_1 for Os should lie between 439(10)GPa and 443(8)GPa, while that (B_2) of Ir between 354(10)GPa and 361(21)GPa (cf. as for the B^d values they were found appreciably smaller than B as expected from the thermodynamical model discussed in the previous Section). In other words, our findings concerning the bulk modulus of Os do not support the claim by *Cynn et al.* (2002) that it is markedly larger than the bulk modulus reported by *Occelli et al.* (2003) for diamond which is 443GPa. As for our findings concerning the bulk modulus B_2 of Ir, we see that it lies between the values 354(6)GPa and 391(4)GPa reported by *Cerenius and Dubrovinsky* (2000) and *Goncharov et al.* (2007), respectively.

4 Summary and Conclusions

Here, we made use of the bulk moduli of fcc- and hcp-structured Ir-Os alloys recently measured by *Yusenko et al.* (2015) in order to estimate the bulk modulus of osmium for which controversial values have been reported. We find that it lies between 439(10)GPa and 443(8)GPa by employing a model different than that used in (*Yusenko et al.*, 2015). Our results strengthen the conclusion that the bulk modulus for Os is slightly smaller in comparison with diamond ($B = 443$ GPa). Furthermore, our findings contradict the earlier claim of *Cynn et al.* (2002) that osmium is less compressible than diamond, which is particularly interesting due to the significance of Os as a possible model of iron behavior in the Earth's core.

Acknowledgments

The experimental data used in the present paper come from Tables 2 and 3 of *Yusenko et al.* (2015).

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